

**PATENT APPLICATION**  
**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of Docket No: Q76524

Yutaka TOSAKI, et al.

Appln. No.: 10/616,916 Group Art Unit: 1713

Confirmation No.: 2409 Examiner: Kelechi Chidi Egwim

Filed: July 11, 2003

For: AQUEOUS DISPERSION TYPE PRESSURE-SENSITIVE ADHESIVE COMPOSITION  
AND PRESSURE-SENSITIVE ADHESIVE PRODUCT

**SUPPLEMENTAL DECLARATION UNDER 37 C.F.R. § 1.132**

Mail Stop Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Yutaka Tosaki, hereby declare and state:

THAT I am a citizen of Japan;

THAT I have received a Bachelor's degree on March 1986 from Kobe University,  
Faculty of Engineering Department of Industrial Chemistry;

THAT I have been employed by Nito Denko Corporation since April of 1986, where I  
have been engaged in research and development relating to pressure-sensitive adhesive tapes to  
date.

I am a co-inventor of the invention described and claimed in the above-identified  
application.

I am familiar with the Office Action dated August 11, 2006. In regard to the rejection of present claims 1 and 3, the following experiments were carried out, either by me or under my direct supervision.

**1) Re: The Rejection over Iijima et al. (Iijima)**

In the claims of Iijima, the weight content of the water-soluble polyol is defined to be from 3 to 25%, and in the examples, one case of 15% by weight is shown. Thus, in the Declaration submitted on July 26, 2006, I conducted a replication experiment faithfully obeying the compositional example. However, in view of the Examiner's position that the present claim language does not exclude a pressure-sensitive composition of Iijima, which contains glycerin in an amount of as little as 3% by weight. Therefore, I conducted an additional replication experiment using the smallest content of 3% by weight of glycerin. The results are shown below.

**1-1) Procedures for sample preparation for replication experiment**

The following three compositions have been reproduced.

**Comparative Experiment 1**

Discussion about the use of the same polymers as in Example 1 of Iijima et al

Material	Polymer	Composition	Manufacturer	Commercial product name
2-ethylhexyl Acrylate	80	89	Toagosei Co.	2EHA
Methyl Methacrylate	20		Mitsubishi Gas Chemical Co.	MMA
Ammonium Lauryl Sulfate	3		Kao Co.	LATEMUL AD-25
Polyvinyl Alcohol		8	Nippon Synthetic Chem. Ind. Co.	Gosenol GM-14

Glycerin		3	Wako Pure Chemical Co.	
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To a reaction vessel equipped with a thermometer, a stirrer, a nitrogen introducing tube and a reflux cooling tube, 40 parts of water were charged, and 300 ml/min of nitrogen was introduced at room temperature for 1 hr. Thereafter the content was heated to 70°C, and 0.4 parts of sodium persulfate dissolved in 10 parts of water were added. Then, a mixture obtained by emulsifying 160 parts of 2-ethylhexyl acrylate (a product of Toagsei Co., Ltd.) and 40 parts of methyl methacrylate (a product of Mitsubishi Gas Chemical Co., Inc.) into 69 parts of water with use of 25 parts of ammonium lauryl sulfate (LATEMUL AD-25, solid content of 24%, a product of Kao Corp.) was added dropwise over the period of 1 hr 40 min. During this addition, the temperature of the inner bath was kept at 70°C. Thereafter, by cooling the content and adjusting its pH neutral by adding 2.4 parts of 10% aqueous ammonia at the inner bath temperature of 30°C, an emulsion of the main polymer was obtained.

To the solid content (89% by weight) of this emulsion, were added 8% by weight of poly(vinyl alcohol) (Gosenol GM-14; a product of Nippon Synthetic Chemical Co., Ltd.) and 3% by weight of glycerin (a product of Wako Pure Chemical Industries, Ltd.) as shown in the table above. The pressure-sensitive adhesive solution thus prepared was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to have a thickness of 20 µm after drying, and dried to obtain a pressure-sensitive adhesive tape.

Comparative Experiment 2

Discussion about the use of the same polymers as in Example of the Present Application

Material	Polymer	Composition	Manufacturer	Commercial product name
2-ethylhexyl Acrylate	92	94	Toagosei Co.	2EHA
Butyl Acrylate	5		Toagosei Co.	BA
Acrylonitrile	2		Mitsui Chemical Co.	AN
Acrylic Acid	1		Toagosei Co.	AA
Ammonium Lauryl Sulfate	3		Kao Co.	LATEMUL AD-25
Polyethylene Glycol		3	Wako Pure Chemical Co.	
Glycerin		3	Wako Pure Chemical Co.	

A main polymer emulsion was obtained in a similar manner as in Comparative Experiment 1 except that the monomer composition in the above table has been adopted.

As shown in the above table, 3% by weight of poly(ethylene glycol) (a product of Wako Pure Chemical Industries, Ltd., with a weight-average molecular weight of 500,000) and 3% by weight of glycerin (a product of Wako Pure Chemical Industries, Ltd.) were added to 94% by weight of the solid content of this emulsion.

The resulting pressure-sensitive adhesive solution was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to give a thickness of 20 µm after drying, and dried to give a pressure-sensitive adhesive tape.

Comparative Experiment 3

Discussion about the use of the quite different polymers

Material	Polymer	Composition	Manufacturer	Commercial product name
Iso-nonyl Acrylate	92	94	Nippon Syokubai Co.	INA
Butyl Methacrylate	5		Mitsubishi Rayon Co.	BMA
Styrene	2		Kishida Chemical Co.	
Methacrylic Acid	1		Mitsubishi Rayon Co.	MAA
Ammonium Lauryl Sulfate	3		Kao Co.	LATEMUL AD-25
Polyethylene Glycol			Wako Pure Chemical Co.	
Glycerin		3	Wako Pure Chemical Co.	

The main polymer emulsion was obtained in a manner similar to Comparative Experiment 1 except adoption of the monomer composition shown in the above table. As shown in the above table, 3% by weight of poly(ethylene glycol) (a product of Wako Pure Chemical Industries, Ltd., with a weight-average molecular weight of 500,000) and 3% by weight of glycerin (a product of Wako Pure Chemical Industries, Ltd.) were added to 94% by weight of the solid content of this emulsion.

The resulting pressure-sensitive adhesive solution was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to give a thickness of 20 µm after drying, and dried to give a pressure-sensitive adhesive tape.

**1-2) Results of the replication experiments**

The results of measuring the wet-surface adhesive forces of the above-described three kinds of pressure-sensitive adhesive tapes by the method as set forth in the working example of the present application are shown in the table below.

	Wet surface adhesive force (N/18-mm width)
Comparative Experiment 1	0.02
Comparative Experiment 2	0.2
Comparative Experiment 3	0.15

The wet-surface adhesive force in the example of the present application has values ranging from 0.5 to 0.6 N/18-mm width, obviously showing that the result of Comparative Experiment 1 gives low values. As a cause for such result, glycerin, i.e., a water-soluble polyol added to each system is regarded as having an adverse effect. Since the water-soluble polyol has a low molecular weight, it is adsorbed on the surface of the emulsion particles, covering the particle surface. Under such conditions, the hydrophilic component (i.e., the water-soluble polyol) is also present on the surface of the pressure-sensitive adhesive. Thus, the moisture owing to dewing on the surface of the adherend will not be adsorbed into the interior of the pressure-sensitive adhesive since it is energetically stabilized at the pressure-sensitive adhesive-adherend interface. In this connection, due to the presence of a water layer at the pressure-sensitive adhesive-adherend interface, the wet-surface adhesive force gives a low value. The same conclusion can be derived from the results of Comparative Experiments 2 and 3 of the present application. On the other hand, only in the case of high-molecular weight poly(alkylene glycol) defined by the present application, the glycol exists in an aggregated dispersed state

without covering the particle surface since it has a high molecular weight (Figure 1). Since these portions are particularly likely to absorb water, and are not present at the surface of the pressure-sensitive adhesive, the moisture due to dewing becomes energetically unstable at the interface between the adherend and the pressure-sensitive adhesive, thus being absorbed into the interior of the pressure-sensitive adhesive. As a result, the water content at the interface between the adherend and the pressure-sensitive adhesive reduces to enhance wet-surface adhesive force.

Figure 1 is a cross-sectional TEM photograph showing the pressure-sensitive adhesive in Example 1 of the present application, in which the portion lying in the lower left side of the top slanted line is the pressure-sensitive adhesive wherein the dark aggregated part consists of poly(ethylene glycol).

As has been discussed heretofore, it is seen that wet-surface adhesive force lowers by adding 3% by weight of a water-soluble polyol as a hydrophilic, low-molecular weight component, indifferent to the type of the polymer component.

2) Re: The rejection over Rosenski et al. (Rosenski)

Concerning the substitution of unavailable materials and repeated replication experiment

2-1) Materials that have been substituted due to unavailability are as follows.

<1> 'Abex26s'

Though Abex26s is described as an alkyl phenol ether sulfate, the length of the alkyl group and the detail of the sulfate salt are not described.

With respect to the emulsifier, as set forth in the part starting with column 4, line 57, there is described 'The emulsifier includes anionic, cationic and nonionic ones that have been

conventionally used in emulsion polymerization . . . . Usually, at least one kind of anionic emulsifier is used, and one or more nonionic emulsifier may be used, too . . . . A specific example is disodium dodecyldiphenyl ether disulfonate . . . . Accordingly, in the replication experiment, an alkylidiphenyl ether disulfonate: PELEX SSH (a product of KAO Co.) was used. In other words, this material is one specifically described in the cited reference.

<2> 'Sodium vinyl sulfonate'

Though sodium vinyl sulfonate is used as a stabilizing comonomer, no description is found in the body text of the specification on 'any stabilizing comonomer and sodium vinyl sulfonate' at all. Thus, instead of the above-described material that is difficult to obtain, sodium allylsulfonate was used. These materials belong to simple vinyl monomers each containing sodium sulfonate, and their fundamental properties are substantially the same.

<3> 'Monoctyl maleate'

In the replication experiment in the Declaration submitted on July 26, 2006, 'dioctyl maleate' was used for the experiment as an alternative of 'monoctyl maleate'. Though 'dioctyl maleate' is a substantially equivalent material to 'monoctyl maleate', it has been found that the same molar number must be incorporated as that of monoctyl maleate in terms of carboxylic group content as a result of confirmation of the invented details of Rosenski. Hence, I conducted an additional replication experiment with the following formulation. Namely, in Example 1, 30.0 g of monoctyl maleate (molecular weight = 228) is used, and the molar number of carboxylic group is  $30.0/228 = 0.13$  mole. Accordingly, acrylic acid (molecular weight = 72) was added in  $0.13 \times 72 = 9.36$  g as an alternative material. Via this addition, the carboxylic

functional value becomes equal to that in Example 1. By way of precaution, acrylic acid as an alternative material for monoctyl maleate is set forth in the detailed description of the cited reference as 'one of the preferable materials as the carboxylic acid monomer'.

#### 2-2) Results of the replication experiment

The formulations of Rosenski are shown in the table below.

Example 1 of Rosenski et al.

	Material	Grams	Manufacturer	Material name and information on physical properties
A	Deionized Water	255		
	Sodium Acetate (buffer)	0.675		
	PBG 8000 (plasticizer)	90	Union Carbide	Polyethylene glycol (Mw8000)
	Aerosol MA (anionic surfactant)	1.5	American Cyanamid	Dihexyl ester of sodium sulfosuccinic acid
	Abex 26S (anionic surfactant)	1.35	Rhone-Poulenc	Alkyl phenol ether sulfate
B	Siponic L4 (nonionic surfactant)	3.0	Rhone-Poulenc	Polyoxyethylene (4) lauryl alcohol
	2-Ethylhexyl Acrylate	24		
C	Vinyl Acetate	6		
D1	Sodium Persulfate	0.3		
	Deionized Water	4.0		
	Deionized Water	66		
D2	Abex 26S	13.8		
	Sodium Vinyl Sulfonate (stabilizing comonomer)	3.0		
	2-Ethylhexyl Acrylate	205.5		
	Vinyl Acetate	45.0		
	Monoctyl Maleate	30.0		
E	Acrylic Acid	1.5		
	Siponic L4	3.0		
	Deionized Water	24		
	Sodium Persulfate (initiator)	0.75		
	Ammonium Hydroxide (neutralizing agent)	9.9		

	Material	Grams	Manufacturer	Material name and information on physical properties
F	t-Butyl Hydroperoxide (scavenger)	0.3		
	Deionized Water	1.0		
G	Sodium Metabisulfite (reducing agent)	0.6		
	Deionized Water	4.5		

Based on the above-described working example, the replication experiment was carried out with the following formulation.

**Replication Experiment for Example 1 of Rosenski et al**

	Material	Grams	Manufacturer	Material name and information on physical properties
A	Deionized Water	253		
	Sodium Acetate (buffer)	0.675	Kishida Chemical Co.	
	PEG 6000 (plasticizer)	90	Kishida Chemical Co.	Polyethylene Glycol (Mw 8000)
	Sodium Dialkyl Sulfosuccinate	2.143	Kao Co.	PELEX OTP 70%
	Sodium Alkyl Diphenyl Ether Disulfonate	2.7	Kao Co.	PELEX SS-H 50%
	Polyoxyethylene Lauryl Ether	3.0	Kao Co.	EMULGEN 104P, 100%
B	2-Ethylhexyl Acrylate	24	Toagosei Co.	
	Vinyl Acetate	6	Denki Kagaku Kogyo Co.	
C	Sodium Persulfate	0.3	Kishida Chemical Co.	
	Deionized Water	4.0		
D1	Deionized Water	52.2		
	Sodium Alkyl Diphenyl Ether Disulfonate	27.6	Kao Co.	PELEX SS-H, 50%
	Sodium Allylsulfonate (stabilizing comonomer)	3.0	Tokyo Kasei Kogyo Co.	
D2	2-Ethylhexyl Acrylate	205.5	Toagosei Co.	
	Vinyl Acetate	45.0	Denki Kagaku Kogyo Co.	
	Acrylic Acid	9.36	Toagosei Co.	Substitute of Monoctyl Maleate

Supplemental Declaration under 37 C.F.R. § 1.132  
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	Material	Grams	Manufacturer	Material name and information on physical properties
	Acrylic Acid	1.5	Toagosei Co.	
	Polyoxyethylene Lauryl Ether	3.0	Kao Co.	EMULGEN 104P, 100%
E	Deionized Water	24		
	Sodium Persulfate (initiator)	0.75	Kishida Chemical Co.	
	Ammonium Hydroxide (neutralizing agent)	9.9	Kishida Chemical Co.	
F	t-Butyl Hydroperoxide (scavenger)	0.429	Kayaku Akzo Co.	KayabutylH70, 70%
	Deionized Water	0.871		
G	Sodium Metabisulfite (reducing agent)	0.6	Kishida Chemical Co.	
	Deionized Water	4.5		

Re: The materials used in Example 1

- As described above, I replaced 'Abex26s', 'sodium vinyl sulfonate' and 'monooctyl maleate' with alternative materials that demonstrate fundamentally the same properties.
- With respect to the other materials, those having the same chemical formulations were used. Thus, I conclude that Example 1 has been faithfully reproduced.
- Some quantity differences in the formulation are the result of re-calculation so that accurate solid contents were employed. This is because the surfactants, etc., are in aqueous solution form.

The polymerization formulation was faithfully reproduced in conformity with Example 1.

The resulting pressure-sensitive adhesive solution was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to give a thickness of 20 µm after drying, and dried to give a pressure-sensitive adhesive tape.

[Evaluation Result]

The result obtained by the measurement of wet-surface adhesive force of the above-described pressure-sensitive adhesive tape according to the method described in the examples of the present application is shown in the following table.

	Wet-surface adhesive force (N/18-mm width)
Example 1 of Rosenski et al.	0.03

As shown in the table above, the wet-surface adhesive force resulted in a low value. The reason is that, since polymerization is conducted under the state where the polyoxyethylene plasticizer is incorporated, the polyoxyethylene plasticizer is adsorbed on the particle surface. And even when the materials are fabricated into a tape, the plasticizer exists in the surface of the pressure-sensitive adhesive, raising the hydrophilic nature of the surface of the pressure-sensitive adhesive, whereby, when the tape is laminated with the wet surface of an adherend, moisture exists between the adherend and the surface of the pressure-sensitive adhesive energetically stably. Namely, the moisture is not absorbed into the interior of the pressure-sensitive adhesive, resulting in a low wet-surface adhesive force.

In my invention, this task is achieved by restricting the lower limit of the hydrophilic polymer to 100,000, and further by adding the polymer after polymerization, thus suppressing the migration of the hydrophilic polymer to the surface of the pressure-sensitive adhesive.

3) Re: The rejection over Kunihiro et al. (Kunihiro)

To verify that the invention of Kunihiro is different from the present application, Example 5 of Kunihiro was reproduced. I chose Example 5 because Example 5 is the only example that uses poly(ethylene glycol), which is the poly(alkylene glycol) defined by the present application (This is the exemplified embodiment closest to the present application.). In addition, the other examples in Kunihiro are expected to basically exhibit the same tendency as Example 5.

The most significant difference between Kunihiro and my invention lies in whether polymerization is conducted in the presence of poly(alkylene glycol), etc. or poly(alkylene glycol) is added after polymerization.

Example 5 of Kunihiro et al.

Material	Part	% by mass	Manufacturer	Material name and information on physical properties
2-ethylhexyl Acrylate	250			
Acrylic Acid	35			
Methyl Methacrylate	20			
Methoxyethyl Acrylate	75			
Vinyl Acetate	20			
Anionic Emulsifier	6		Kao Co.	Emal NC-35
Ion Exchanged Water	150			
Polyethylene Glycol		0.9	Wako Pure Chemical Co.	
Ion Exchanged Water	150			
Potassium Persulfate	3			
Octyl Mercaptan	3			
Triethanol Amine	50 % by mole of carboxyl groups			

**Formulation for Replication Experiment**

Example 5 of Kunihiro et al.				Material name and information on physical properties
Material	Part	% by mass	Manufacturer	
2-ethylhexyl Acrylate	250	100	Toagosei Co.	2EHA
Acrylic Acid	35		Toagosei Co.	AA
Methyl Methacrylate	20		Mitsubishi Gas Chemical Co.	MMA
Methoxyethyl Acrylate	75		Toagosei Co.	ACRICS C-1
Vinyl Acetate	20		Denki Kagaku Kogyo Co.	
Anionic Emulsifier	6		Kao Co.	Emal NC-35
Ion Exchanged Water	150			
Polyethylene Glycol	3.6	0.9'	Wako Pure Chemical Co.	PEG1000
Ion Exchanged Water	150			
Potassium Persulfate	3		Kishida Chemical Co.	
Octyl Mercaptan	3		Kishida Chemical Co.	
Triethanol Amine	36.26		Kishida Chemical Co.	

In the examples of Kunihiro, 1/6 of the emulsified monomer mixture is first added dropwise, and when the reaction ratio reaches 90%, the remaining monomer mixture is added dropwise in 3 hours. Therefore, in advance of conducting the present polymerization, 1/6 of an emulsified monomer mixture having the same formulation was added dropwise, and the time required for achieving the reaction ratio of 90% was derived.

The results are as shown below, wherein the reaction ratio of 90% was obtained in 120 min.

Elapsed time	0 min	15 min	30 min	60 min	90 min	120 min
Reaction ratio (%)	0	53.1	72.7	82.7	85.6	90.7

Therefore, in the present polymerization, 1/6 of the emulsified monomer mixture was first added dropwise, and then after 120 min, the remaining monomer mixture was added dropwise over the period of 3 hours for conducting polymerization. It is further described that the amount of triethanolamine, which is added for the purpose of neutralization, etc. after polymerization, is 50% by mole of the carboxylic group in the copolymer. However, in the examples, the actual added amount is not stated. In Example 5, the monomer containing a carboxylic group is only acrylic acid, whose molar number is  $35/72 = 0.486$  mole. Then, the amount of triethanolamine (molecular weight = 149.19) to achieve 1/2 mole is  $0.486 \times 0.5 \times 149.19 = 36.26$ . The other compositional amounts and polymerization formulation were reproduced faithfully to those of Example 5.

The resulting pressure-sensitive adhesive solution was coated on one surface of a Japanese paper base material with a basis weight of 30 g/m<sup>2</sup> so as to give a thickness of 20 µm after drying, and dried to give a pressure-sensitive adhesive tape.

#### [Evaluation Results]

The result obtained by the measurement of wet-surface adhesive force of the above-described pressure-sensitive adhesive tape according to the method described in the examples of the present application is shown in the following table.

	Wet-surface adhesive force (N/18-mm width)
Example 1 of Kunihiro et al.	0.04

As shown in the table above, the wet-surface adhesive force resulted in a low value. The reason is that, since polymerization is conducted under the state where polyethylene glycol is incorporated, the polyethylene glycol is adsorbed on the particle surface. And even when the materials are fabricated into a tape, the glycol exists also in the surface of the pressure-sensitive adhesive, raising the hydrophilic nature of the surface of the pressure-sensitive adhesive, whereby, when the tape is applied to the wet surface of an adherend, moisture exists between the adherend and the surface of the pressure-sensitive adhesive energetically stably. Namely, the moisture is not absorbed into the interior of the pressure-sensitive adhesive, and it is considered that a low wet-surface adhesive force resulted. This is the same result as in the cited reference on Rosenski, and I consider the case to be the mechanistic demerit of the hydrophilic polymer incorporated prior to polymerization. Moreover, the fact that the molecular weight is as low as 1,000 is not preferable for wet-surface adhesive force, too.

In my invention, the task is achieved by restricting the lower limit of the hydrophilic polymer to 100,000, and further by adding the polymer after polymerization, thus suppressing the migration of the hydrophilic polymer to the surface of the pressure-sensitive adhesive.

In addition, Kunihiro states in Claim 1 'copolymers obtained via copolymerization in the presence of at least one of '(poly)alkylene glycol di(meth)acrylate, (poly)alkylene glycol mono(meth)acrylate, (poly)ethylene glycol, and (poly)propylene glycol'. Among them,

however, the former two are reactive monomers, while the latter two are non-reactive additives. It is not easy for one of ordinary skill in the art to arrive at the (poly)alkylene glycol that has the structure as well as the molecular weight defined in my invention from such a group in which reactive monomers and additives are mixed together, and thus it is concluded that the present application is an utterly new invention. With respect to the adhesive force to wet-surfaces or the constant load peeling property relating to the tasks of the present application, Kunihiro has no description at all, indicating that the purpose as well as the design concept is completely different between the two inventions.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: January 9, 2007

Yutaka Tosaki  
Yutaka Tosaki

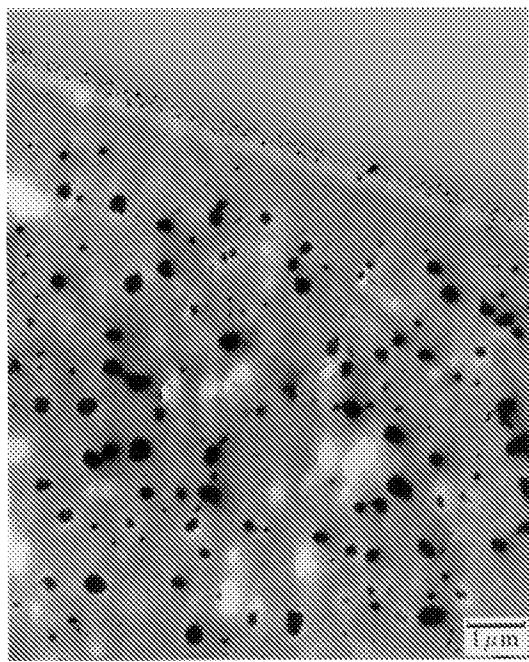


Fig. 1